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Effect of Introducing Receptive Layer to Paper Substrate in Powder Electroluminescent Device

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Abstract

A powder electroluminescent (EL) device is a flat light-emitting device producible only via printing process. The properties of the EL device can be improved by introducing a receptive layer into the paper substrate surface, although the underlying mechanisms of this enhancement remain unknown. Herein, this mechanism was investigated in terms of dielectric loss. The tan δ of the device prepared with the receptive layer was high compared to that obtained without the receptive layer. The slopes of the current and luminance steeply increased accompanying a decrease of tan δ. Therefore, this frequency property determines the frequency properties of the EL device.

1. Introduction

Inorganic electroluminescence is defined as the emission of light from an inorganic material such as a ZnS-type phosphor induced by accelerating an electron under a high electric field. In general, this phenomenon is utilized in back light devices such as watches and mobile phones. In particular, powder EL devices can reduce material costs, time, and equipment required to fabricate the devices compared to semiconductor process because they are fabricated via printing process. Furthermore, devices containing an Ag nanowire electrode exhibit high bending stability, as reported by Park et al. [1]. Recently, devices have been developed that can emit light under voltage application, generate sound [2], generate electric power due to triboelectric charging by structural changes [3], and recover their structure due to the addition of self-recovering materials such as polyacrylic acid hydrogel and polyurethane [4]. Moreover, these powder EL devices have attracted significant attention from academic and industrial researchers. They are expected to facilitate the production of versatile light-emitting devices for illumination and digital signage.

In conventional substrates, glass and plastic films are used. From an environmental perspective, top emission devices prepared using a paper substrate have been reported by Kim et al. [5]. The paper substrate is light, inexpensive, and workable compared to conventional substrates. However, the surface asperity of the paper substrate strongly affects the current properties of the device. To overcome this issue, Kim et al. reported the planarization of a paper substrate in a EL device, significantly improving EL properties [5]. In, powder EL devices, the capacitance has a close relationship with luminance; however, the underlying mechanisms of these enhancements remain unclear. Additionally, when an AC voltage is applied, a dielectric loss dependent on the capacitance occurs, and electrical energy is lost as thermal energy. Therefore, in powder EL devices on paper substrates, the mechanisms of EL enhancements can likely be determined by analyzing the dielectric loss.

This study was performed to analyze the mechanisms of the abovementioned EL enhancements in a powder EL device with a bottom emission structure from which luminescence was obtained on the substrate side.

Herein, the mechanism was examined by measurement of the frequency dependence of the current, luminance, and tan δ in the bottom emission devices. In addition, each layer of the powder EL devices were reported as equivalent series circuits, in resistance and capacitor [6-8]. Therefore, the powder EL devices herein were calculated as a RC equivalent series circuit.

2. Experimental

2.1 Materials

Tracing paper as substrate was purchased from SAKAE Technical-paper Co., Ltd. Zinc sulfide (ZnS)-type particles (GG45) as a phosphor layer were purchased from OSRAM SYLVANIA. Barium titanium oxide ($BaTiO_3$) as a dielectric layer was purchased from KISHIDA CHEMICAL Co., Ltd. Cyanoresin (CR-V) as a high dielectric polymer was purchased from SHINETSU Chemical Co., Ltd. Cyclohexanone as a solvent was purchased from FUJIFILM Wako Pure Chemical Co. Poly (3,4-ethylenedioxythiophene) -poly (styrenesulfonate) (PEDOT: PSS) as a transparent electrode was purchased from SIGMA-ALDRICH Co., LLC. Ag-paste as a back electrode was purchased from MINO GROUP Co., Ltd. All materials were used without further purification. Material derived from cellulose was used as a receptive layer to planarize the paper substrate because it is easy to print on paper.

2.2 Preparation of the EL devices

The high-dielectric polymer paste was prepared by mixing cyanoresin and cyclohexanone at a weight ratio of 3:7. The phosphor particle and $BaTiO_3$ were dispersed in the high-dielectric polymer paste at a weight ratio of 4:6. These functional materials were laminated using an automatic screen printing machine (TU2020-C: SERITECH Co., Ltd.). Each material was sintered in a constant-temperature oven (FS-405: Yamato Scientific Co., Ltd.) at 150 °C for 6 min. The transparent electrode was sintered for 40 min.

2.3 Measurements

The frequency dependences of the current and luminance of the prepared devices were measured using an EL measurement system (SX-1152: IWATSU ELECTRIC CO., Ltd.). The frequency dependences of tan δ, capacitance and equivalent series resistance of the devices were measured using an LCZ-meter (4276A: Agilent Technologies Japan, Ltd.).

3. Results and Discussion

3.1 EL properties of the prepared devices

To examine the effect of the receptive layer on the powder EL device, the frequency dependences of the current and luminance of the devices were measured. Fig. 1 shows the frequency dependence of the currents under ±300 V AC voltage. Upon introduction of the receptive layer to the paper substrate, the current of the powder EL device increased to 17 mA, 1.2 times higher than that of the powder EL device without a receptive layer (14 mA). This was likely due to the decreased surface resistivity by planarization of the paper substrate. The increased current with increasing frequency was non-linear in these devices. To determine the distance of the current values in Fig. 1, the delta of each current was obtained (Fig. 2). In the device without a receptive layer, ΔI increased with increasing frequency, marked by a peak at 8 kHz. Furthermore, the device with a receptive layer exhibited similar behavior to that prepared without a receptive layer, but the peak was observed at a lower frequency. To evaluate the relationship between the current and luminance, the frequency dependence of the luminance was measured in the two devices at a voltage of ±300 V AC (Fig. 3). The increase in luminance with increasing frequency was non-linear, similar to the current behavior. Thus, the increased of luminance corresponded to increasing current. Moreover, upon introduction of a receptive layer to the paper substrate, the luminance of the powder EL device increased to 2200 cd/m^2, 1.4 times higher than that of the powder EL device prepared without a receptive layer (1600 cd/m^2). Therefore, the receptive layer strongly affected the frequency dependence of the luminance compared to the current. This is likely because the phosphor layer was applied, resulting in a uniform electric field by uniformity of the distance between electrodes.

3.2 Dielectric loss

To further investigate the trend in current values shown in Fig. 1, tan δ was measured for the prepared devices (Fig. 4). In the device without a receptive layer, tan δ decreased with increasing frequency to 11 kHz. In contrast, tan δ of the device with a receptive layer decreased until 7 kHz. Over almost the entire frequency region tested herein, tan δ of the device with the receptive layer was high than that without the receptive layer. When tan δ decreased, the current increased (Figs. 1 and 2). Therefore, this frequency property governed the frequency properties of the EL device. The relationships between tan δ, capacitance, and equivalent series resistance can be expressed by Eq. (1) [9]. Moreover, the decreased tan δ with increasing frequency was similar to that observed in a previous report [7]. Therefore, it is considered that the frequency dependence of capacitance and equivalent series resistance strongly affected tan δ.

$$\tan \delta = 2\pi f C R_S \quad (1)$$

3.3 Capacitance

To analyze the frequency dependence of tan δ in detail, the frequency dependences of the capacitance and equivalent series resistance for the devices were measured. Fig. 5 shows the frequency dependence of the capacitances under a ±1 V effective voltage. Under application of a 500 Hz AC voltage, upon introduction of a receptive layer to the paper substrate, the capacitance of the powder EL device increased to 0.660 nF, 1.3 times higher than that of the powder EL device without a receptive layer (0.498 nF). Thus, it is considered that the distance between electrodes in the device was evened by the introduction of a receptive layer. Furthermore, this result indicates a decreased capacitive reactance in the powder EL device. For this reason, it is likely that the enhanced current was achieved by decreasing the capacitive reactance due to the increased of capacitance (Fig. 1).

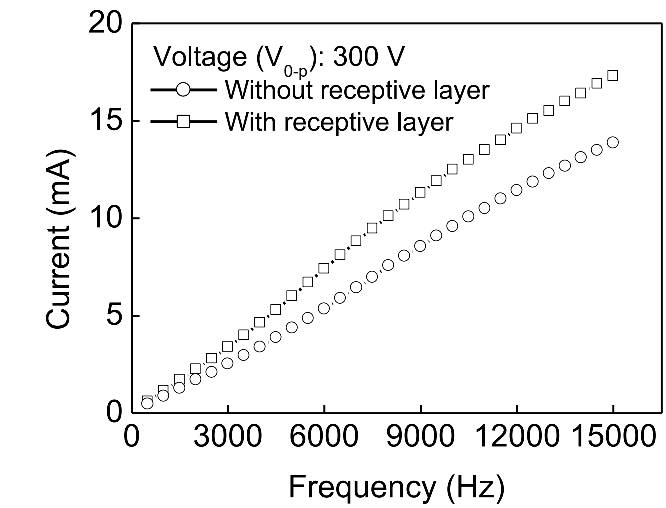

Fig. 1 I-F characteristics at 300 V of the powder EL devices on a paper substrate.

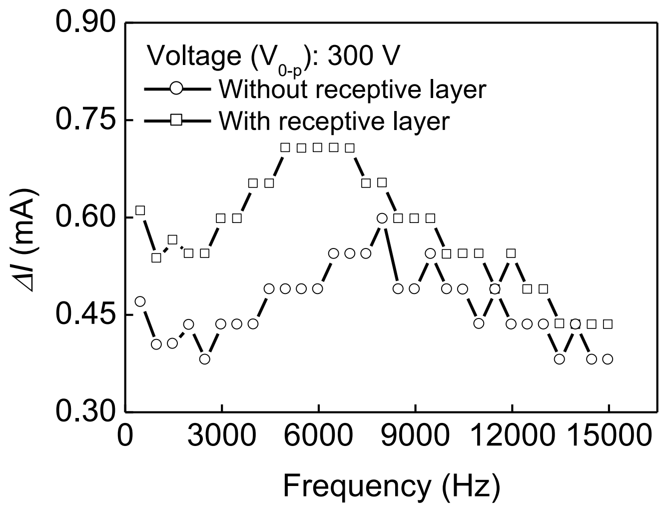

Fig. 2 Current increases and I-F characteristics.

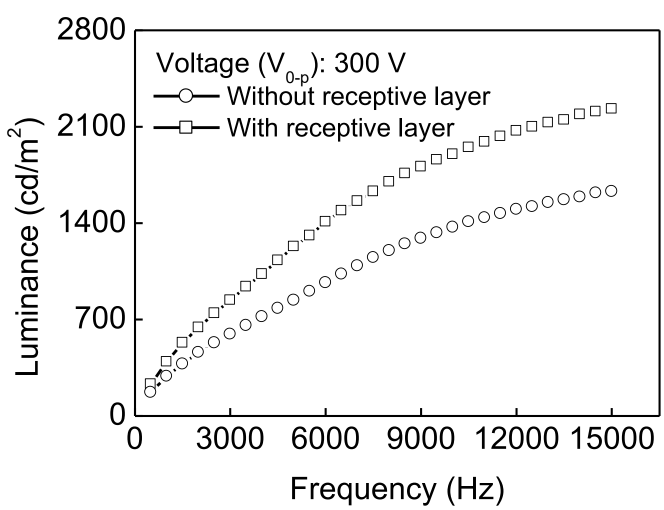

Fig. 3 L-F characteristics at 300 V of the powder EL devices on a paper substrate.

3.4 Equivalent series resistance

The equivalent series resistance under a ±1 V effective voltage decreased with increasing frequency, similar to that of capacitance (Fig. 6). Upon application of a 500 Hz AC voltage, with a receptive layer in the paper substrate, the capacitance of the powder EL device decreased to 93.1 kΩ, lower than that of the powder EL device without a receptive layer (125 kΩ). In the both devices, the equivalent series resistance accompanying the increased frequency was significantly decreased compared to capacitance. Moreover, the equivalent series resistance is important parameter of heat generation, as explained by Obara et al. [10]. Therefore, it is likely that the decreased equivalent series resistance decreased the heat energy generated in the device. Therefore, tan δ was strongly affected by the equivalent series resistance.

4. Conclusion

Herein, to determine the underlying mechanisms of the EL enhancements in powder EL devices by introduction of a receptive layer to paper substrate, the dielectric loss was analyzed. The capacitance and capacitive reactance were improved upon introduction of a receptive layer to the paper substrate, leading to enhanced EL properties in the device. However, the increased current and luminance accompanying the increasing frequency was non-linear in these devices. Furthermore, the increased current actually decreased in frequency region in which tan δ increased. Therefore, it was shown that the EL properties of the devices were affected by electrical energy losses due to the dielectric loss. Moreover, the frequency dependence of tan δ was strongly dependent on the equivalent series resistance due to heat generation. From these results, the frequency properties of the equivalent series resistance greatly affected to the frequency properties of the current and luminance of the powder EL devices. These results are part of the standard evaluation concerning the enhanced of luminance of powder EL devices.

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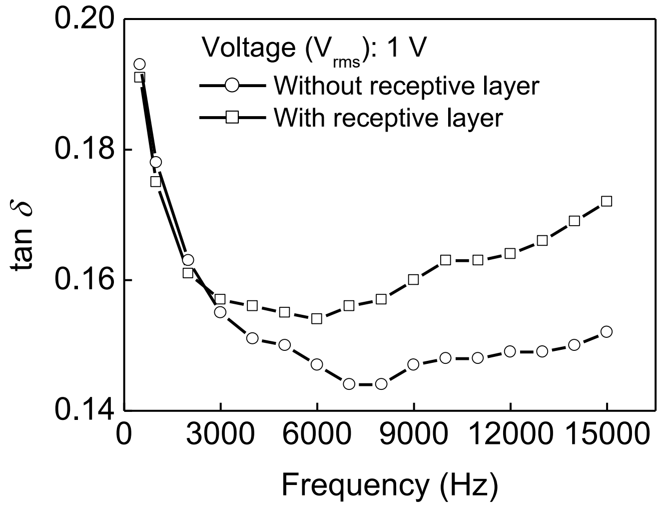

Fig. 4 Dielectric tangent (tan δ) at 1 V of the powder EL devices on a paper substrate.

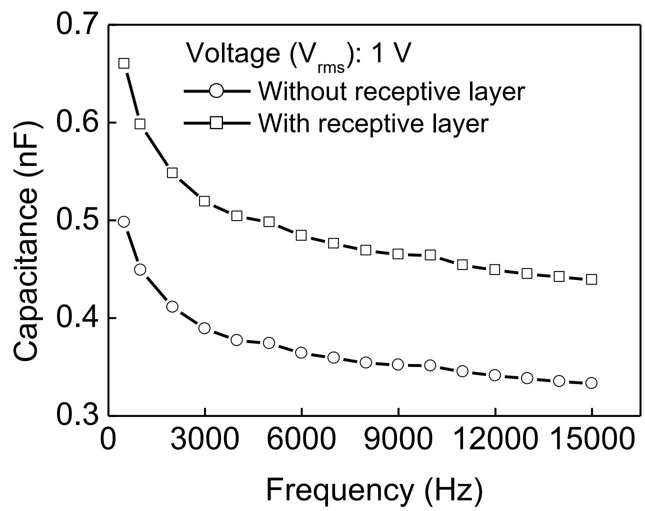

Fig. 5 C-F characteristics at 1 V of the powder EL devices on a paper substrate.

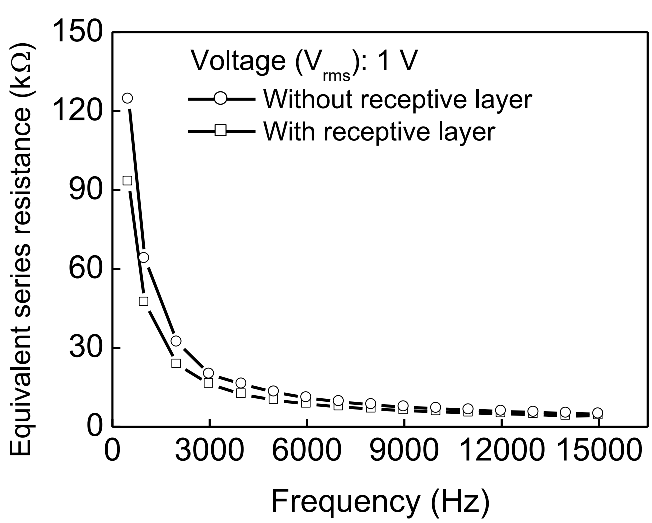

Fig. 6 R_S-F characteristics at 1 V of the powder EL devices on a paper substrate.

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